

PATENT APPLN. NO. 10/576,299  
RESPONSE UNDER 37 C.F.R. § 1.116

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REMARKS

Applicants acknowledge with gratitude the courteous and helpful interview granted to their undersigned representative on December 3, 2008, by Examiners Nwaonicha and Parsa. The substance of the interview is explained below.

In the Action, claims 1-8 and 10 are rejected under 35 U.S.C. 103(a) as being unpatentable over Oono et al., U.S. Patent No. 6,723,483 ("Oono"), in view of Osawa et al., U.S. Patent No. 5,824,824 ("Osawa"), "for the reason stated in the previous Office Action dated 12/04/2007." (Action, page 2, line 3 of the first full paragraph).

In the response filed March 17, 2008, to the first Action, applicants argued that the combination of Oono and Osawa is insufficient to support a case of prima facie obviousness of the claims of the present application under 35 U.S.C. § 103(a) and that, notwithstanding such insufficiency, the comparative data in the application demonstrate unexpected results sufficient to rebut any prima facie obviousness alleged by the Office to be supported by the prior art.

Applicants explained that the present invention, as precisely recited in the claims as amended in the response, relates to a method for producing a triarylsulfonium salt having a structure in

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which only one of the three aromatic rings on the cation portion of the salt is different from the other two aromatic rings (see paragraph [0024] in the present US publication). In the method of the present invention, an activator with high affinity for oxygen is used in an amount of 4.5 to 7.5 equivalents relative to the diaryl sulfoxide reactant.

In the method of the present invention for producing a triarylsulfonium salt [4], a diaryl sulfoxide [1] and an aryl Grignard reagent [2] are reacted in the presence of an activator with high affinity for oxygen (hereinafter, abbreviated as "the activator") in an amount of 4.5 to 7.5 equivalents relative to the the diaryl sulfoxide, and then the resultant reaction mixture is reacted with a strong acid [3], or a salt thereof.

The method of the present invention shows unexpected excellent effects with the use a larger amount of the activator than has been conventionally used. In particular, the use of 4.5 to 7.5 eq. of the activator can obtain a desired sulfonium salt efficiently in a high purity without byproducts (see paragraphs [0033] and [0070] in the US publication). This result could not have been reasonably predicted from the prior art.

More specifically, the method of the present invention has been made to solve the problem that when a diaryl sulfoxide [two

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aromatic rings have the same structure (structure [a])) and an aryl Grignard reagent having an aromatic ring different from the aromatic ring of the diarylsulfoxide (structure [b]) are reacted not only the desired compound (wherein two aromatic rings are structure [a] and one aromatic ring is structure [b]) is obtained, but also two kinds of byproduct are obtained. One byproduct is a sulfonium salt wherein three aromatic rings are all structure [a], and the other byproduct is a sulfonium salt wherein one aromatic ring is structure [a] and two aromatic rings are structure [b]). The present invention has been completed on the basis of the present inventors' finding that use of the activator of 4.5 to 7.5 eq. relative to the diaryl sulfoxide produces the desired sulfonium salt effectively in a high yield without any such byproducts (see paragraph [0007] in the US Publication).

The fact that the desired sulfonium salt can be efficiently produced in a high purity without byproducts is extremely important.

First, when the triarylsulfonium salt not containing byproducts obtained by the method of the present invention is used as a photo acid-generating agent in a photolithography step in the manufacture of a semiconductor, an improvement of roughness on a profile or a sidewall of a hyperfine pattern and formation of a

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good rectangle pattern of reduced edge roughness (see paragraph [0078] in the US Patent Application Publication of the present application, No. 2007/0083060) is obtained.

In contrast, when by-products are contained in a product, use of such a product as an acid generator changes the efficiency of the acid generation. In other words, sensitivity (e.g., acid generation efficiency) to exposure is different between the product (objective compound) and by-products because the structure of the product and by-products is different. And when a product containing by-products, in which by-product content undergoes a change with each production lot, is used as an acid generator, it is difficult to quantify the amount of acid generation.

As a result, formation of a resist pattern with the use of such an acid generator causes problems such as poor-reproducibility of the pattern.

Formation of a hyperfine pattern is leading-edge technology. Therefore, for formation of a resist pattern with good reproducibility, use of a product with high purity as an acid generator is required.

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Second, separation of by-products from the desired triarylsulfonium salt is difficult because the structure of the by-products is similar to that of the desired product.

The separation of the by-product from the desired product requires the use of a preparative liquid chromatography. However, a preparative liquid chromatography is a separation and refinement means normally used on a laboratory scale. It is difficult to use preparative liquid chromatography as a separation means on an industrial-scale due to a limited amount of one preparation isolation.

The obtaining of the desired triarylsulfonium salt without by-products in the method of the present invention by using an activator of 4.5 to 7.5 eq. is unexpected. This is demonstrated by the comparative data in the examples of the present application. In particular, on page 15, Table 7, in the present US publication there is a description of yields of the obtained objective compound and those of byproducts obtained when using chlorotrimethylsilane (TMSCl) as the activator in amounts of 2.5, 3.0, 4.0, 5.0, 6.0, 7.0, 7.5 eq., relative to the diphenyl sulfoxide.

As is clear from the results of Table 7, it can be understood that use of TMSCl as the activator in an amount of 2.5 eq. relative to the diphenyl sulfoxide forms not only byproducts but also a low

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yield of the objective compound (59%) (see Comparative example 1). Further, as is obvious from the results of Experimental Examples 1 and 2, it can be understood that use of TMSCl (activator) in an amount of 3 and 4 eq. can produce the objective compound with a certain amount of yield (72%), but still forms byproduct.

In contrast, as is obvious from the results of Experimental Examples 3 to 6, use of TMSCl (activator) in an amount of 5, 6 and 7.5 eq. relative to the diphenyl sulfoxide according to the method of the present invention does not result in the formation of any byproducts and produces the objective compound with a high yield.

The fact that the desired triarylsulfonium salt can be produced without byproducts cannot be reasonably predicted from Oono or Osawa or from the combination of Oono and Osawa.

First, with respect to Oono, the method of Oono for producing a sulfonium salt also produces a salt whose cation portion has three identical aromatic rings.

Further, an amount of activator to be used in Oono is 0.8 to 2 mol relative to 1 mole of the diarylsulfoxide (0.8 to 2 eq. relative to the diaryl sulfoxide), which is different from that of the activator of the present invention (4.5 to 7.5 eq. relative to the diarylsulfoxide).

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In the Examples of Oono, there is a disclosure of a method for producing a sulfonium salt, where the cation portion has two identical aromatic rings (structure [a]) and one aromatic ring (structure [b]) different from the other two (structure [a]), using a trimethylsilyl sulfonate such as trimethylsilyl triflate in an amount of 0.13 mol relative to diphenyl sulfoxide of 0.1 mol (i.e., an amount of the activator of 1.3 eq. relative to the diphenyl sulfoxide). However, the yield of the sulfonium salt is low (43%) and, as it is clear from the data in the present application as explained above, the method in the example of Oono forms byproducts other than the desired compound.

With respect to Osawa, Osawa discloses a method for producing a triarylsulfonium salt by reacting a diaryl sulfoxide and an aryl Grignard reagent in the presence of trimethylsilyl chloride (activator) of 1 to 5 mole, preferably 2 to 3 mole, relative to the diaryl sulfoxide (see column 11, line 32, to column 12, line 4). However, the sulfonium salt obtained by Osawa's method includes a sulfonium salt having three identical aromatic rings of the cation portion. In contrast, as has been explained previously, the sulfonium salt obtained by the method of the present invention is limited to a sulfonium salt having one aromatic ring different from

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the others of the three aromatic rings of the cation portion. Therefore, Osawa differs from the present invention in this point.

Moreover, although Osawa includes examples of synthesis of a sulfonium salt having two identical aromatic rings (structure [a]) and one aromatic ring different from the other two (structure [b]), in these examples, TMSCl of 2.5 or 3 eq., relative to the diaryl sulfoxide (see column 17, synthetic example 1; column 18, synthetic example 2; and column 20, synthetic example 4) is used and there is no disclosure relating to byproducts.

During the interview with Mr. Nwaonicha and Mr. Parsa, the comparative data were discussed. The Examiners agreed to reconsider the comparative data in the application and, particularly, the data relating to the amount of by-product produced in the prior art processes. Mr. Parsa suggested that comments be included in the response explaining the difficulty of purifying a product containing by-product.

However, it appeared the Examiners were unsure whether the comparative data in the specification of the present application could overcome the prior art since Osawa discloses the use of an amount of activator of 1-5 eq., which overlaps the range of the amount of activator used in the method of the present invention.



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Applicants respectfully submit that it is clear that obviousness resulting from overlapping ranges can be overcome by a showing of the criticality of the claimed range. (See MPEP § 2144.05(III)). The comparative data in the application demonstrate criticality for the use of the claimed range of amount of activator of 4.5-7.5 eq. The data show that no byproduct is produced with the use of this range of activator. On the other hand, the data show that the use of an amount of activator outside this range and, particularly, an amount of activator as disclosed by Oono and Osawa, results in the formation of undesired byproduct that, as explained above, cannot be easily removed and adversely affects the performance of the product when used as a photo acid generating agent.

For these reasons, removal of the 35 U.S.C. § 103(a) rejection is in order and is respectfully solicited.

The present Action also includes an objection to the specification of the present application relating to the inclusion of Japanese text in the publication of the present application. As explained during the interview, the inclusion of Japanese text in the publication of the present application is not a proper basis for an objection to the present specification. The present specification itself does not include any Japanese text.

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The foregoing is believed to be a complete and proper response to the Office Action dated July 21, 2008.

In the event that this paper is not considered to be timely filed, applicants hereby petition for an appropriate extension of time. The fee for any such extension may be charged to Deposit Account No. 111833.

In the event any additional fees are required, please also charge Deposit Account No. 111833.

Respectfully submitted,

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